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8. PERFORMING ORGANIZATION REPORT

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5 Pollux Drive
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13. SUPPLEMENTARY NOTES

14. ABSTRACT

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2303 M2C8

MEMORANDUM FOR PRS (In-House/Contractor Publication)

FROM: PROI (STINFO) 17 May 2002

SUBJECT: Authorization for Release of Technical Information, Control Number: **AFRL-PR-ED-VG-2002-118**
Jerry Boatz (PRSP) et al., "New Materials Design" (Viewgraphs)

DoD Users Group Conference **(Statement A)**
(Austin, TX, 10-14 June 2002) **(Deadline: 07 June 2002)**

1. This request has been reviewed by the Foreign Disclosure Office for: a.) appropriateness of distribution statement, b.) military/national critical technology, c.) export controls or distribution restrictions, d.) appropriateness for release to a foreign nation, and e.) technical sensitivity and/or economic sensitivity.
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Signature _____ Date _____

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3. This request has been reviewed by the STINFO for: a.) changes if approved as amended, b) appropriateness of references, if applicable; and c.) format and completion of meeting clearance form if required
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Comments:_____

APPROVED/APPROVED AS AMENDED/DISAPPROVED

_____

PHILIP A. KESSEL Date
Technical Advisor
Space and Missile Propulsion Division

New Materials Design

DoD UGC, 10-14 Jun 02

Austin, TX

Jerry Boatz

Senior Research Chemist

Propulsion Directorate

Air Force Research Laboratory

NEW MATERIALS DESIGN

THE TEAM....

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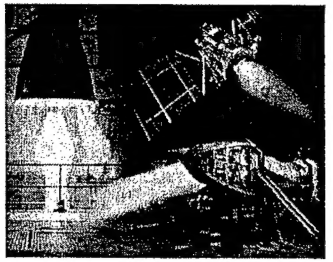
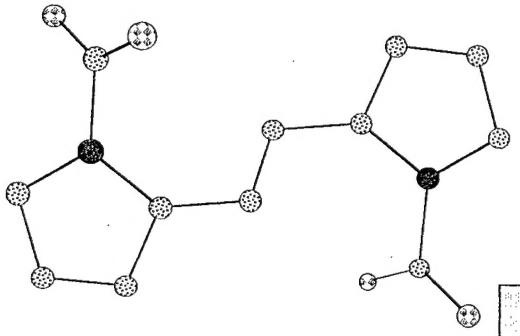


OUTLINE

1. **Project Overview**
   - High energy density matter
   - Polyhedral oligomeric silsesquioxanes (POSS)
   - Non-linear optical materials
2. **Theoretical Methods and benchmarks**
   - Ab initio electronic structure theory
   - Nuclear-electronic orbital approach
   - Centroid Molecular Dynamics
3. **Results**
4. **Summary**

PROJECT OVERVIEW - HEDM

High Energy Density Matter -- next generation rocket propellants

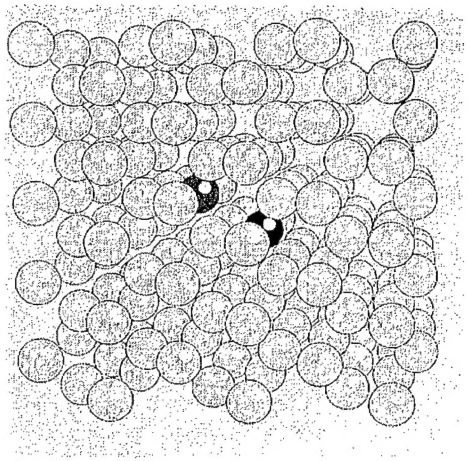


High-nitrogen/polynitrogen compounds

Specific Impulse

$$I_{sp} \propto \sqrt{\Delta H / \overline{m}}$$

Atom-doped solid hydrogen



PROJECT OVERVIEW - HEDM

Technical issues being addressed using CCM

1. High-nitrogen/polynitrogen compounds

Objective: identify, characterize, and synthesize stable compounds with high heats of formation, high densities

- structures, energy content, stabilities, reaction pathways

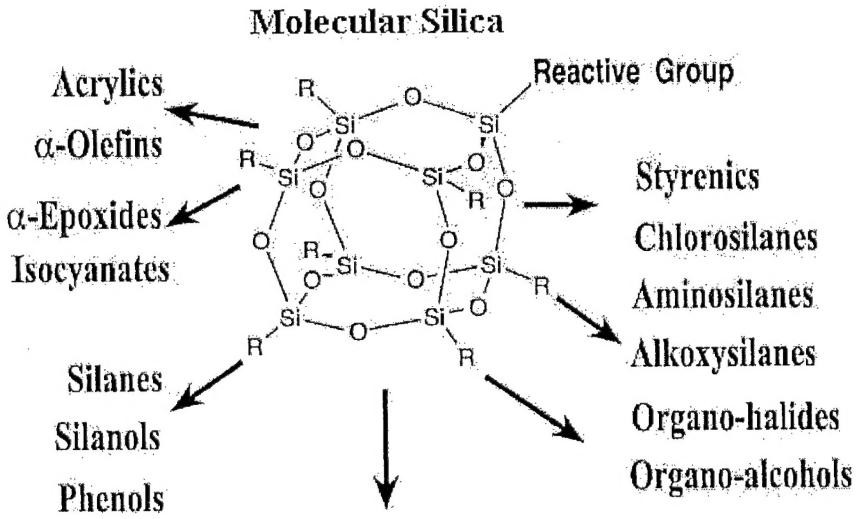
2. Energetic atoms in solid hydrogen

Objective: stabilize ~5% energetic atoms in solid hydrogen

- stabilities, mobilities, concentration limits of atoms stored in hydrogen matrices

PROJECT OVERVIEW - POSS

Polyhedral oligomeric silsesquioxanes -- next generation plastics



As Additives

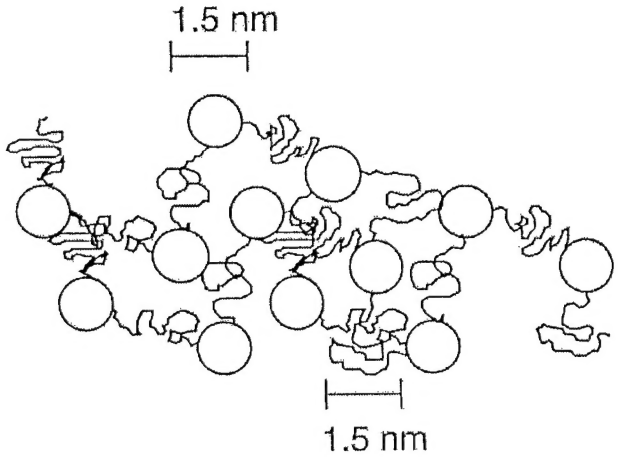
Heat/abrasion resistant paints and coatings
Mechanical property/viscosity/thermal modifiers
Crosslinking agents
Fire retardants

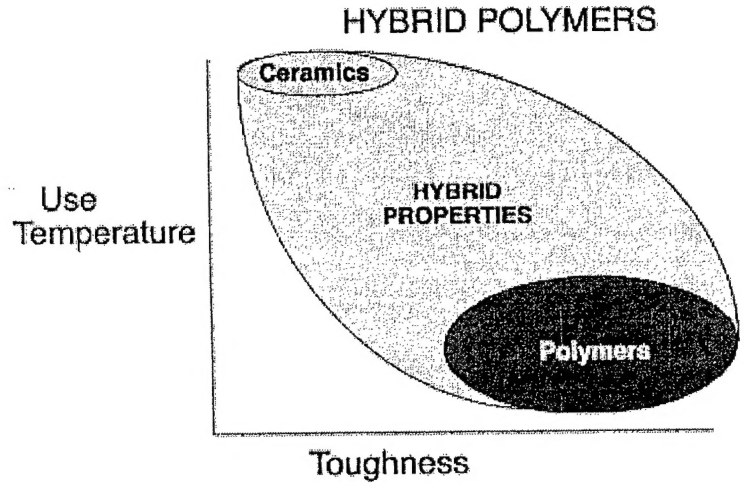
As Plastics

Medical materials
Space resistant resins
Packaging/coatings
Electronic materials
Optical Plastics

As Preceramics

Ablative materials (nozzles, insulations etc.)
Claddings/electronics coatings
Precursors to glassy or ceramic matrices





PROJECT OVERVIEW - POSS

Technical issues being addressed using CCM

1. Mechanisms of formation

Objective: rational design and synthesis of POSS

- role of solvents, acid/base catalysis, substituent effects on mechanism of formation

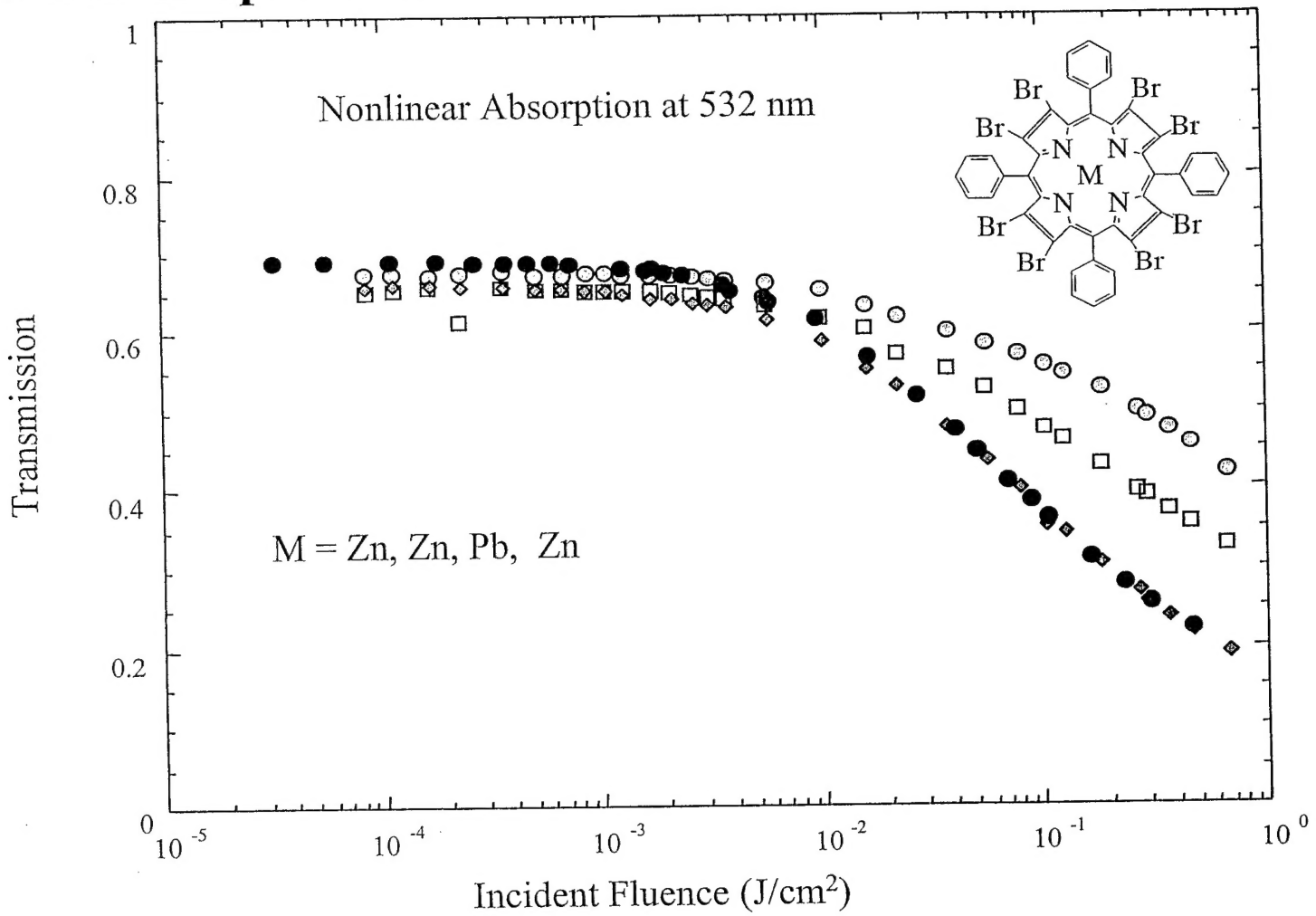
2. Potential applications as molecular "sieves"

Objective: determine if POSS cages can be used to separate small molecules

- determine barriers to encapsulation of N_2 and O_2

PROJECT OVERVIEW - NLO

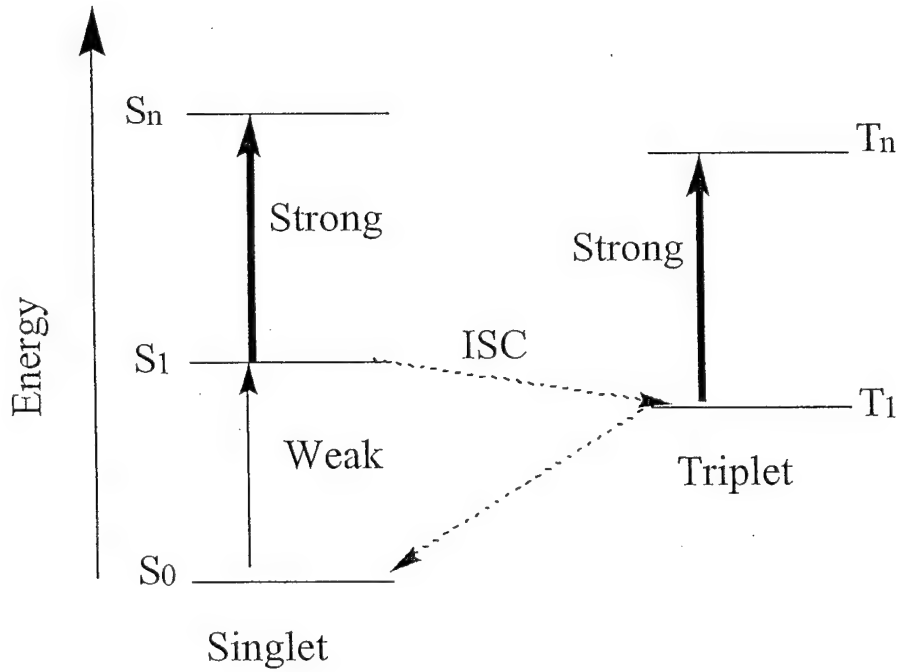
Non-linear optical materials for laser-hardened applications



PROJECT OVERVIEW - NLO

Technical issues being addressed using CCM

1. Mechanism of reverse saturable absorption (RSA)



Five-level model for nonlinear absorption

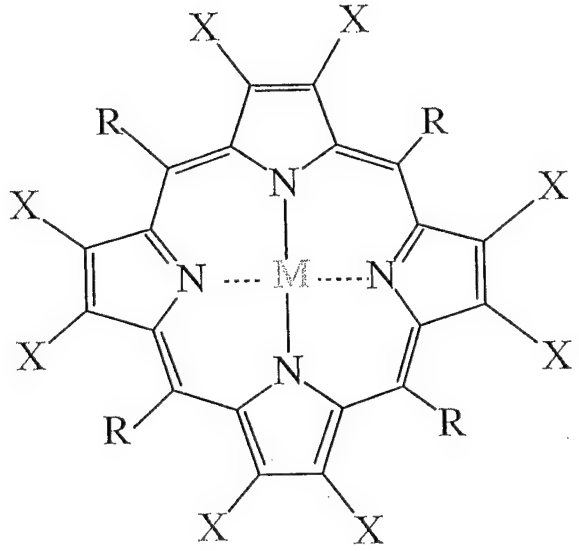
Key Components of RSA

- **Ground state spectra (linear response)**
- **Singlet-triplet splittings (ISC, phosphorescence)**
- **Ionization potential (photoionization)**
- **Excited triplet spectra (NLA)**

PROJECT OVERVIEW - NLO

Technical issues being addressed using CCM

2. "Tuning" of absorption spectrum by benzannulation, halide substitution



System				Property			
	M	X	R	IP	S_0-S_n	S_0-T_1	T_1-T_n
PH_2	H_2	H	H	E, C	E, C	E, C	E, C
ZnP	Zn	H	H	E, C	E, C	E, C	E, C
$TPPH_2$	Zn	H	ϕ	E, C	E, C	E, C	E, C
ZnTPP	Zn	H	ϕ	E, C	E, C	E, C	E, C
$ZnTPPBr_8$	Zn	Br	ϕ	C	E, C	E, C	E, C

IP = Ionization Potential, S_0 -S_n = Ground State Spectrum,
S_0 -T_1 = Singlet-Triplet Gap T_1 -T_n = Triplet-Triplet Spectrum
E = Experiment, C = Calculated

THEORETICAL METHODS

1. **Ab initio electronic structure theory**

   - General Atomic and Molecular Electronic Structure System (GAMESS) -- a CHSSI code

   - Nuclear-electronic orbital approach (NEO) for including nuclear quantum effects (important, e.g., in proton transfer reactions)

Various computational techniques are employed to solve the molecular electronic Schrödinger equation from quantum mechanics:

$$\left[-\frac{1}{2}\sum_i \nabla_i^2 - \sum_i \sum_\alpha \frac{Z_\alpha}{r_{i\alpha}} + \sum_i \sum_{j>i} \frac{1}{r_{ij}}\right]\Psi_{el} = E_{el}\Psi_{el}$$

Categories of approximate solutions:

a) "Self-consistent field" (SCF): reasonably good geometries
b) "Electron correlation": post-SCF correction, required for reliable energetics (e.g., barriers).

THEORETICAL METHODS

1. **Ab initio electronic structure theory (cont.)**

- Most electronic structure codes use Born-Oppenheimer (i.e., "clampled nuclei") approximation -- NOE method treats specified nuclei at QM level.

Nuclear-Electronic Hamiltonian

$$
\begin{aligned}
H_{\mathrm{tot}}(\mathbf{r}_e, \mathbf{r}_q; \mathbf{r}_c) \;=\; & -\sum_i^{N_e} \frac{1}{2}\nabla_i^2 - \sum_i^{N_e}\sum_A^{N_c} \frac{Z_A}{r_{iA}} + \sum_i^{N_e}\sum_{j>i}^{N_e} \frac{1}{r_{ij}} \\
& - \sum_I^{N_p} \frac{1}{2M_I}\nabla_I^2 + \sum_I^{N_p}\sum_A^{N_c} \frac{Z_A Z_I}{r_{IA}} + \sum_I^{N_p}\sum_{J>I}^{N_p} \frac{Z_I Z_J}{r_{IJ}} \\
& - \sum_i^{N_e}\sum_I^{N_p} \frac{Z_I}{r_{iI}} + \sum_A^{N_c}\sum_{B>A}^{N_c} \frac{Z_A Z_B}{r_{AB}}
\end{aligned}
$$

N_e: number of electrons (coordinates $\mathbf{r}_e$)
N_p: number of quantum nuclei (coordinates $\mathbf{r}_p$)
N_c: number of classical nuclei (coordinates $\mathbf{r}_c$)

THEORETICAL METHODS

Ab initio electronic structure theory

Current Status of parallel GAMESS

	RHF	ROHF	UHF	GVB	MCSCF
Energy	cdp	cdp	cdp	cdp	cdp
Analytic Gradient	cdp	cdp	cdp	cdp	cdp
Numeric Hessian	cdp	cdp	cdp	cdp	cdp
Analytic Hessian	cdp	cdp	-	cdp	-
MP2 energy	cdp	cdp	cdp	-	c p
MP2 gradient	cdp	-	cd	-	-
CI energy	cdp	cdp	-	cdp	cdp
CI gradient	cd	-	-	-	-
DFT energy	cdp	cdp	cdp	-	-
DFT gradient	cdp	cdp	cdp	-	-

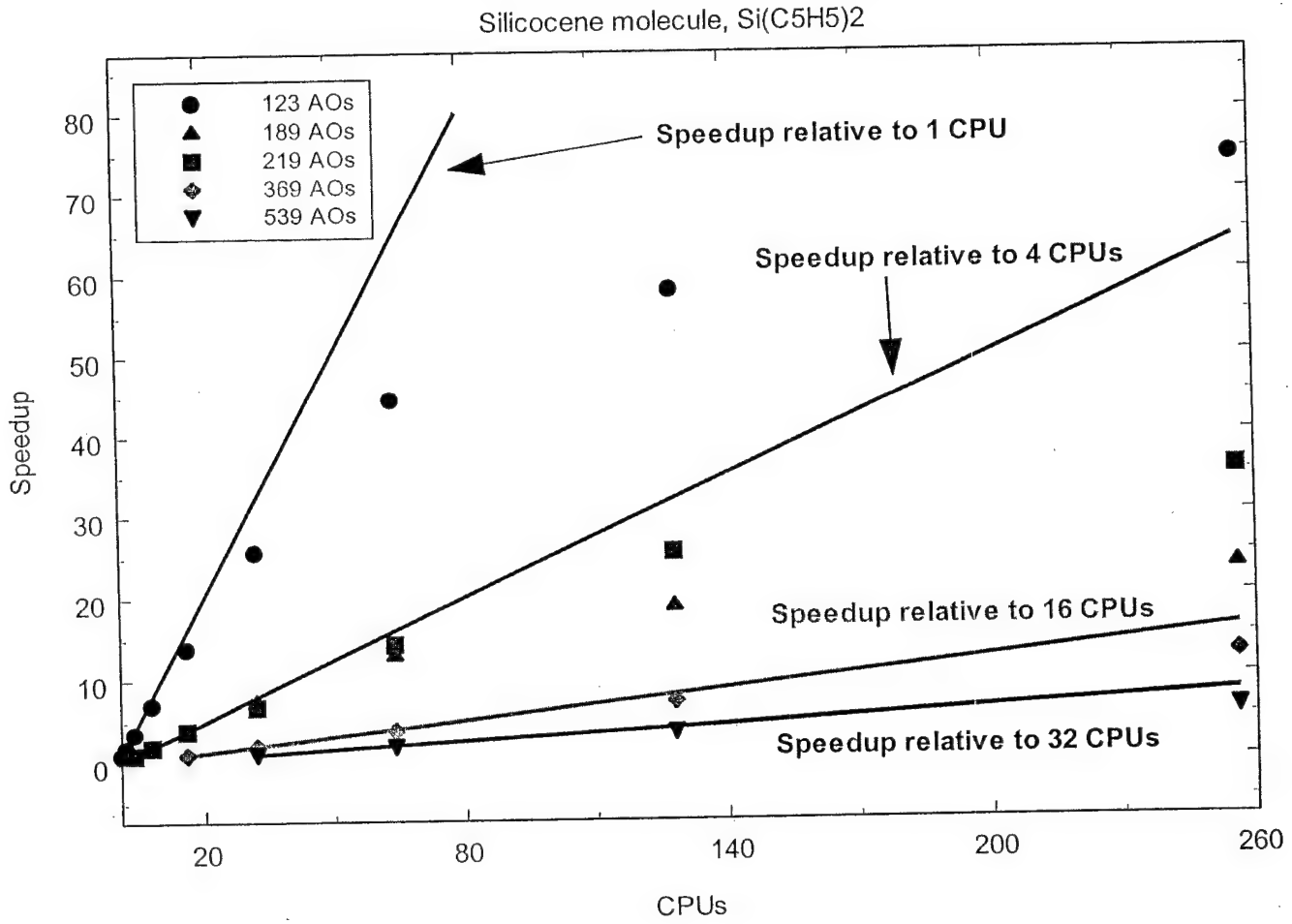
c = conventional disk storage of AO integrals

d = direct evaluation of AO integrals

p = runs in parallel

THEORETICAL METHODS

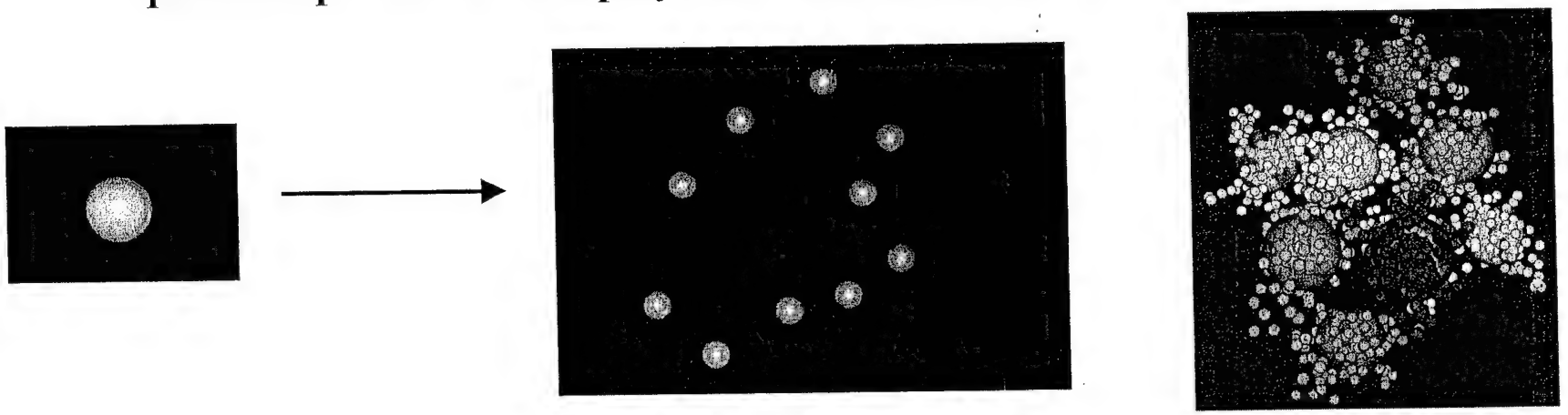
Ab initio electronic structure theory



THEORETICAL METHODS

Path Integral Molecular Dynamics & Centroid Molecular Dynamics (CHSSI codes)

Simulation methods based on path integral techniques for mapping quantum particles onto "polymer ring" of classical quasiparticles:

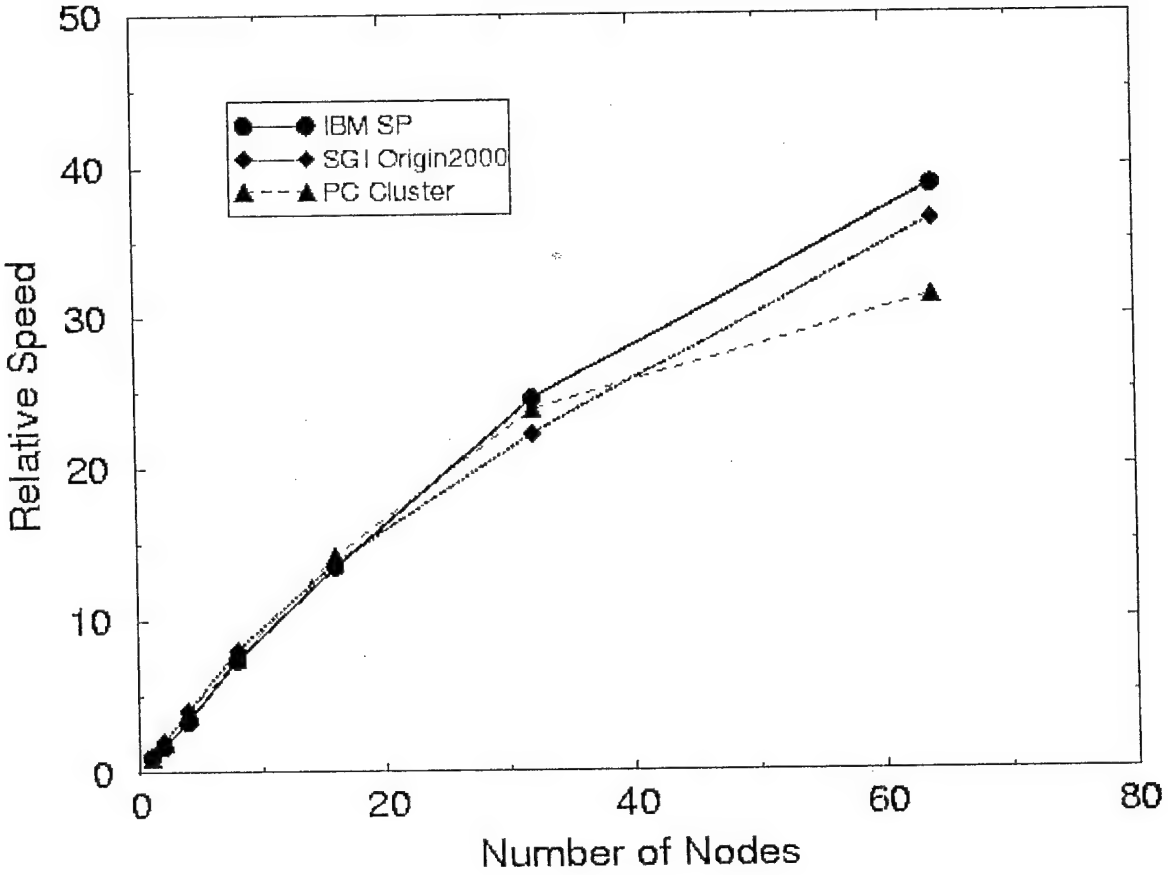


Each "real" particle is replaced by N (50<N<500) quasiparticles; classical dynamics done on collection of quasiparticles => natural, efficient parallelism.

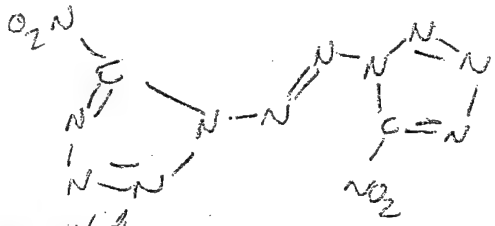
THEORETICAL METHODS

Path Integral Molecular Dynamics & Centroid Molecular Dynamics

Code Scaling



Input>rocket, chamber, 20.410000, exhaust, 0.010000



ΔH_f = +1500 kJ/mol SRI stuff

Rocket specific impulse calculation:

The chamber pressure = 20.41 atm
The exhaust pressure = 0.01 atm
The initial equation error was huge: 24728.149173

The Chamber State:

Reference state = reactants
H(R) = H-1773.07, E(R) = E-1773.05, S(R) = S- 0.00

	P (ATM)	V (CC/GM)	T (K)	H(R) (CAL/GM)	E(R) (CAL/GM)	S(R) (CAL/K/GM)	VGS (CC/GM)
1.)	20.4	909.6888	6436.6	0.00	-449.64	2.389	909.6888

Product concentrations

Name		(mol/kg)	(mol gas/mol explosive)
n2	Gas	2.112e+001	5.410e+000
co	Gas	7.734e+000	1.981e+000
no	Gas	4.609e+000	1.180e+000
o2	Gas	1.561e+000	3.998e-001
co2	Gas	7.513e-002	1.924e-002
no2	Gas	1.418e-003	3.632e-004
*c	solid	0.000e+000	0.000e+000
Total	Gas	3.510e+001	8.990e+000
Total	Cond.	0.000e+000	0.000e+000

The Exhaust State:

Reference state = reactants
H(R) = H-1773.07, E(R) = E-1773.05, S(R) = S- 0.00

	P (ATM)	V (CC/GM)	T (K)	H(R) (CAL/GM)	E(R) (CAL/GM)	S(R) (CAL/K/GM)	VGS (CC/GM)
1.)	0.0	565350.1813	2148.9	-1863.53	-2000.43	2.389	565350.1813

Product concentrations

Name		(mol/kg)	(mol gas/mol explosive)
n2	Gas	2.337e+001	5.984e+000
co2	Gas	6.157e+000	1.577e+000
co	Gas	1.652e+000	4.232e-001
o2	Gas	7.645e-001	1.958e-001
no	Gas	1.234e-001	3.160e-002
no2	Gas	5.283e-006	1.353e-006
*c	solid	0.000e+000	0.000e+000
Total	Gas	3.206e+001	8.212e+000
Total	Cond.	0.000e+000	0.000e+000

The specific impulse = 402.54 seconds

RESULTS - HEDM

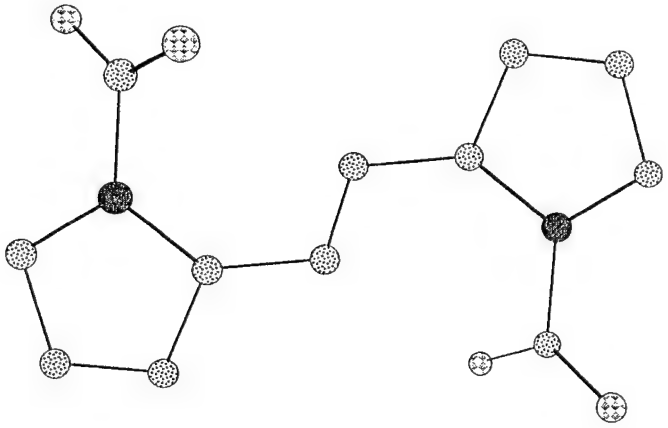
High-nitrogen/polynitrogen compounds

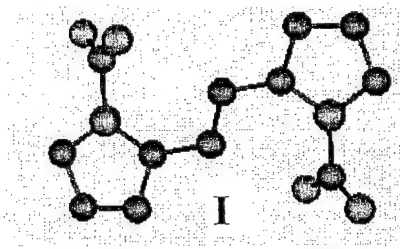
Predicted ΔH_f = 457 kcal/mol, I_{sp} = 329 sec (sea level)

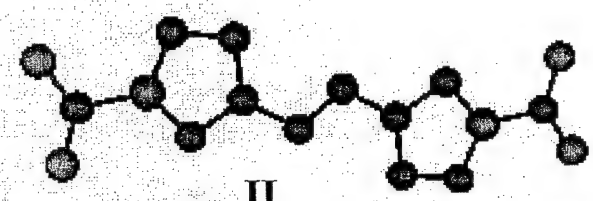
(I_{sp} for hydrazine = 233 sec)

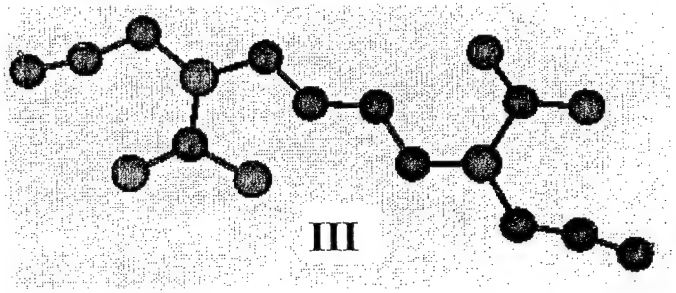
Relative energies (kcal/mol)

I: 0 **II**: -15 **III**: +36



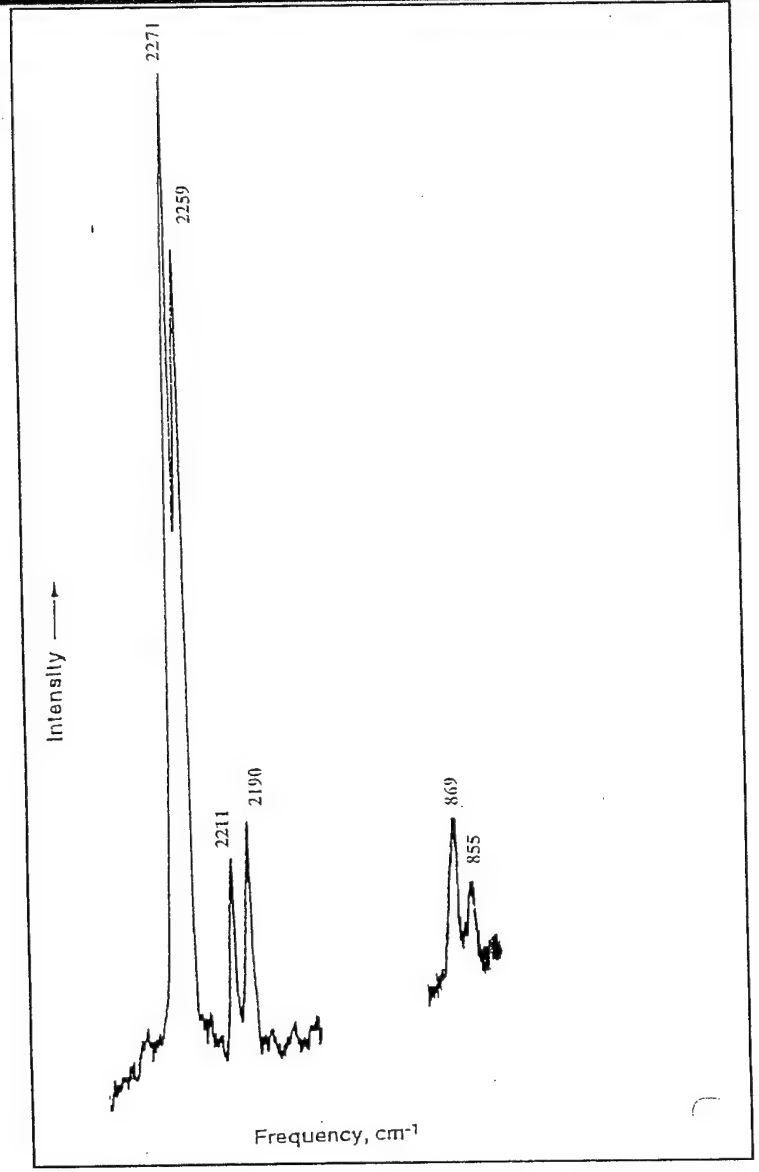


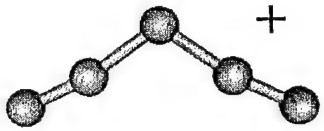




Computational requirements: 4500 MW memory, ~350,000 node-hrs, ERDC T3E

Identifying a Completely New Molecule: Comparison of Calculated and Measured Spectra





$[^{15}N–^{14}N–^{14}N–^{14}N–^{14}N]^{+}AsF_6^{-}$

and

$[^{14}N–^{14}N–^{15}N–^{14}N–^{14}N]^{+}AsF_6^{-}$

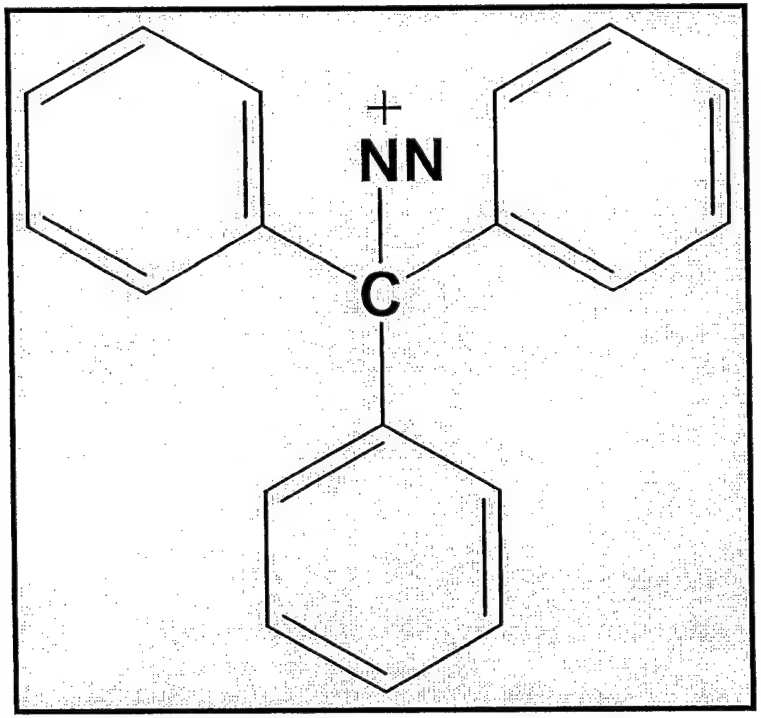
^{14}N - ^{15}N Isotopic Shifts (cm^{-1})

Mode	Obs.	Calc.†
$\nu_1(a_1)$	12	11.8
$\nu_7(b_2)$	21	21.4
$\nu_2(a_1)$	14	14.1

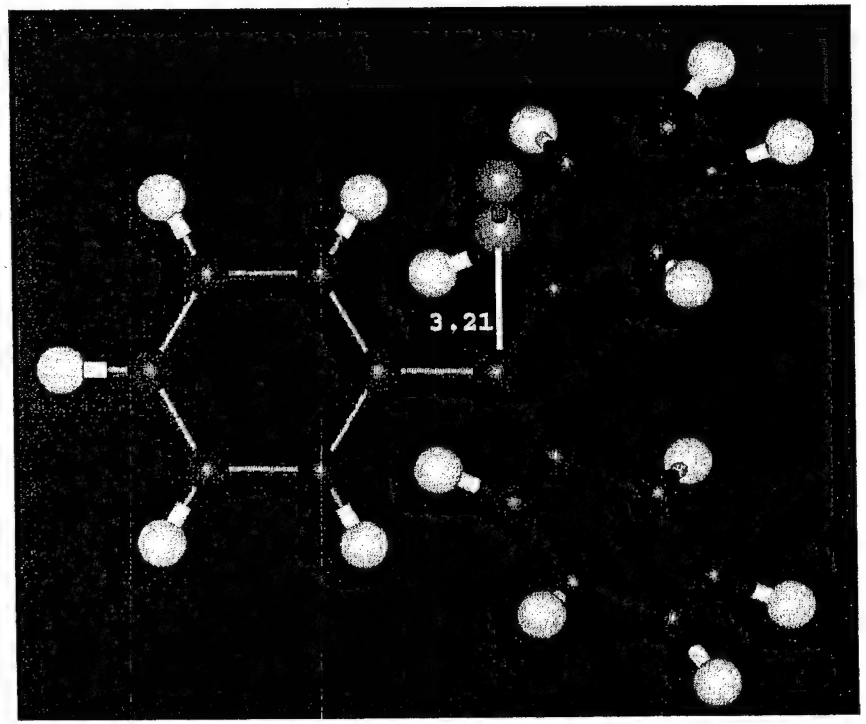
†CCSD(T)/6-311+G(2*d*) results

Identifying Precursors for New Polynitrogens

This ion has been suggested as a useful precursor to new polynitrogen molecules...

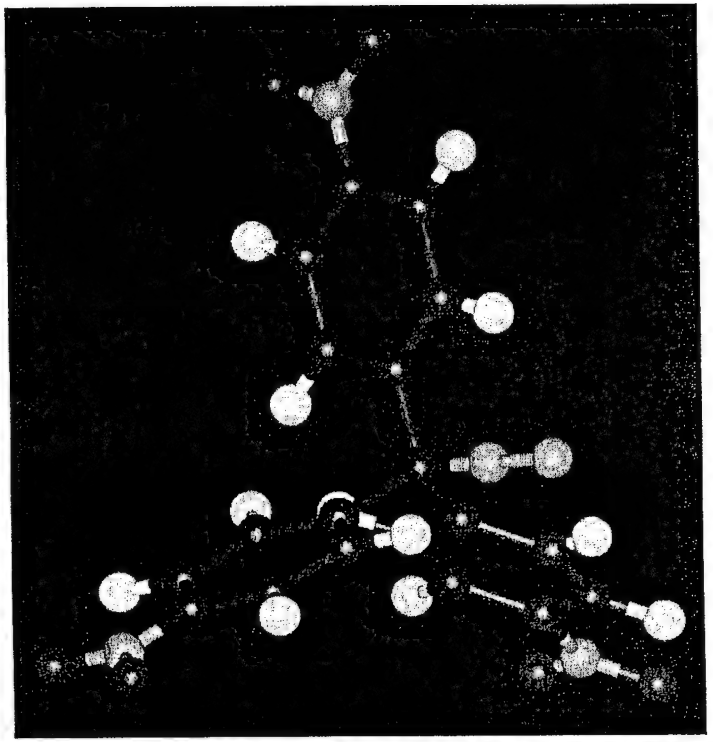


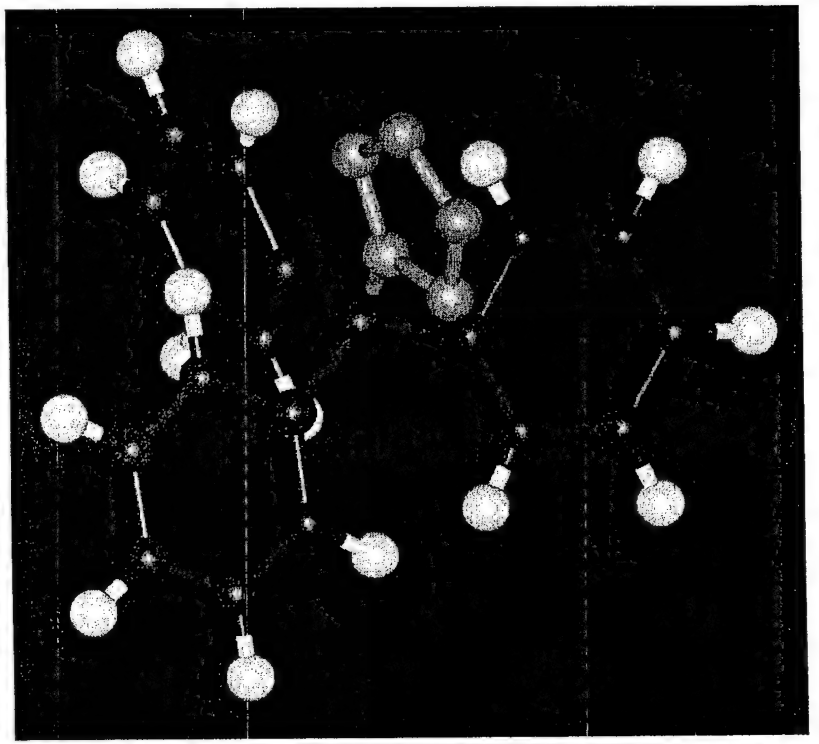
... but calculations predict it to be unstable.

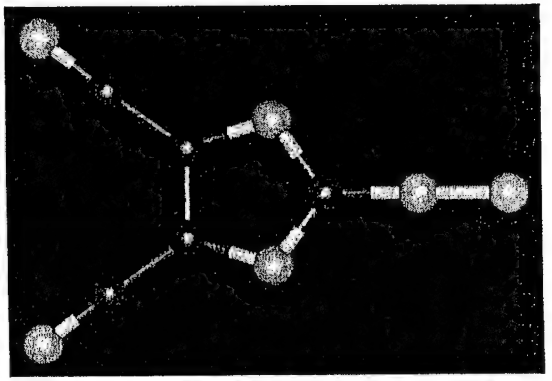


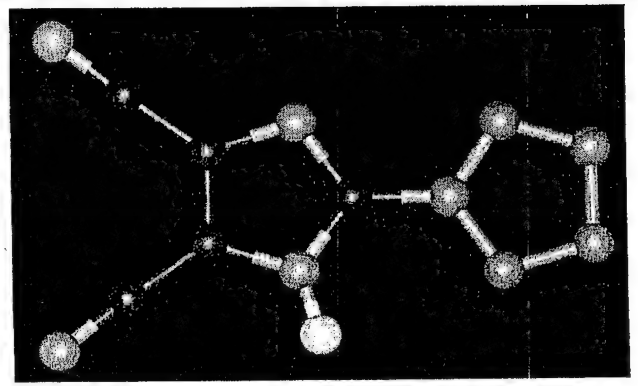
Computational requirements: ~10,000 CPU-hours, 1200 MW on IBM SP/P3 at ASC

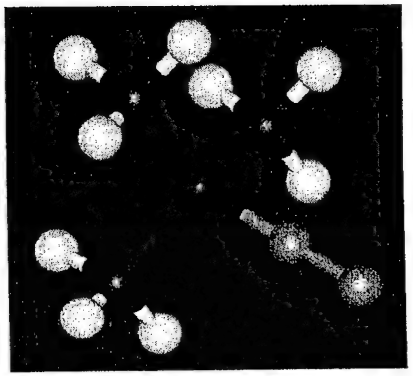
Other Potential Polynitrogen Precursors Being Investigated





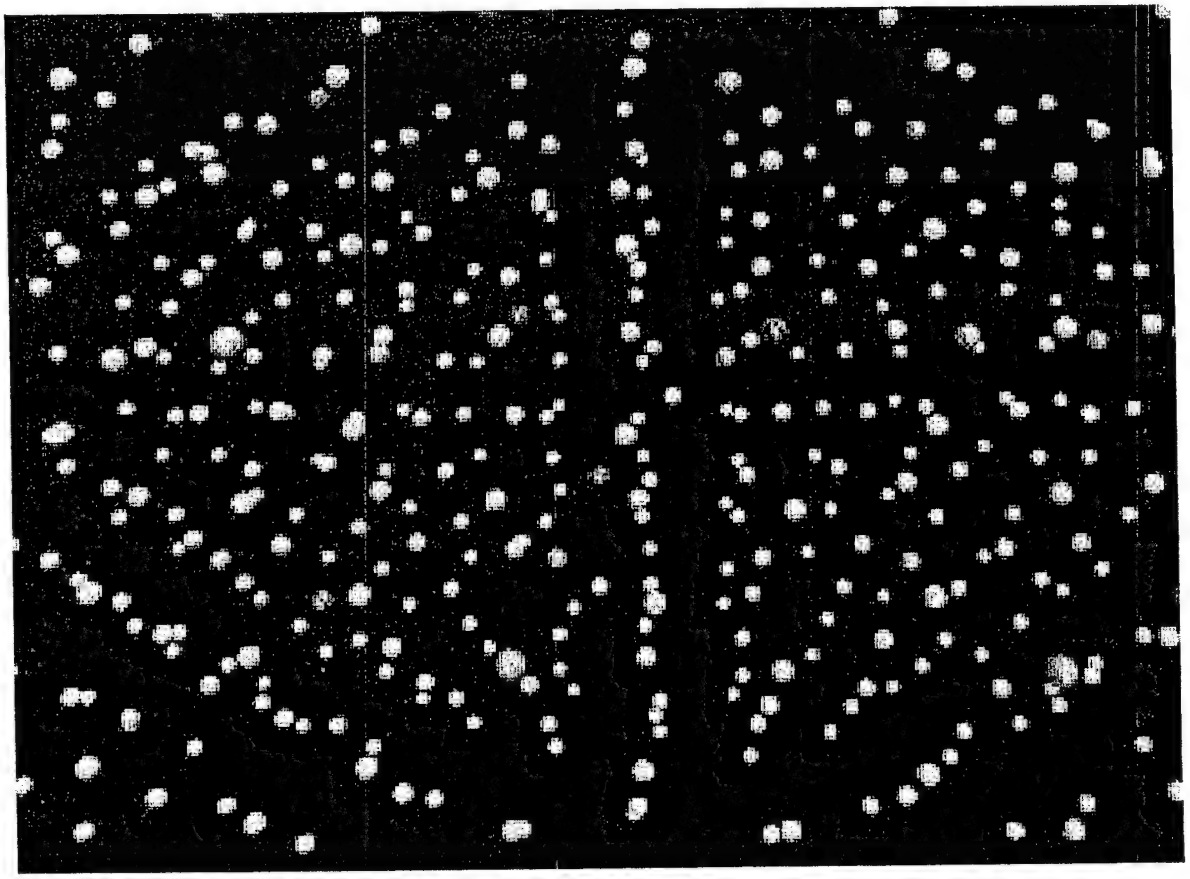






Atom-Doped Solid Hydrogen

6.25% B atoms in solid *para*-H_2



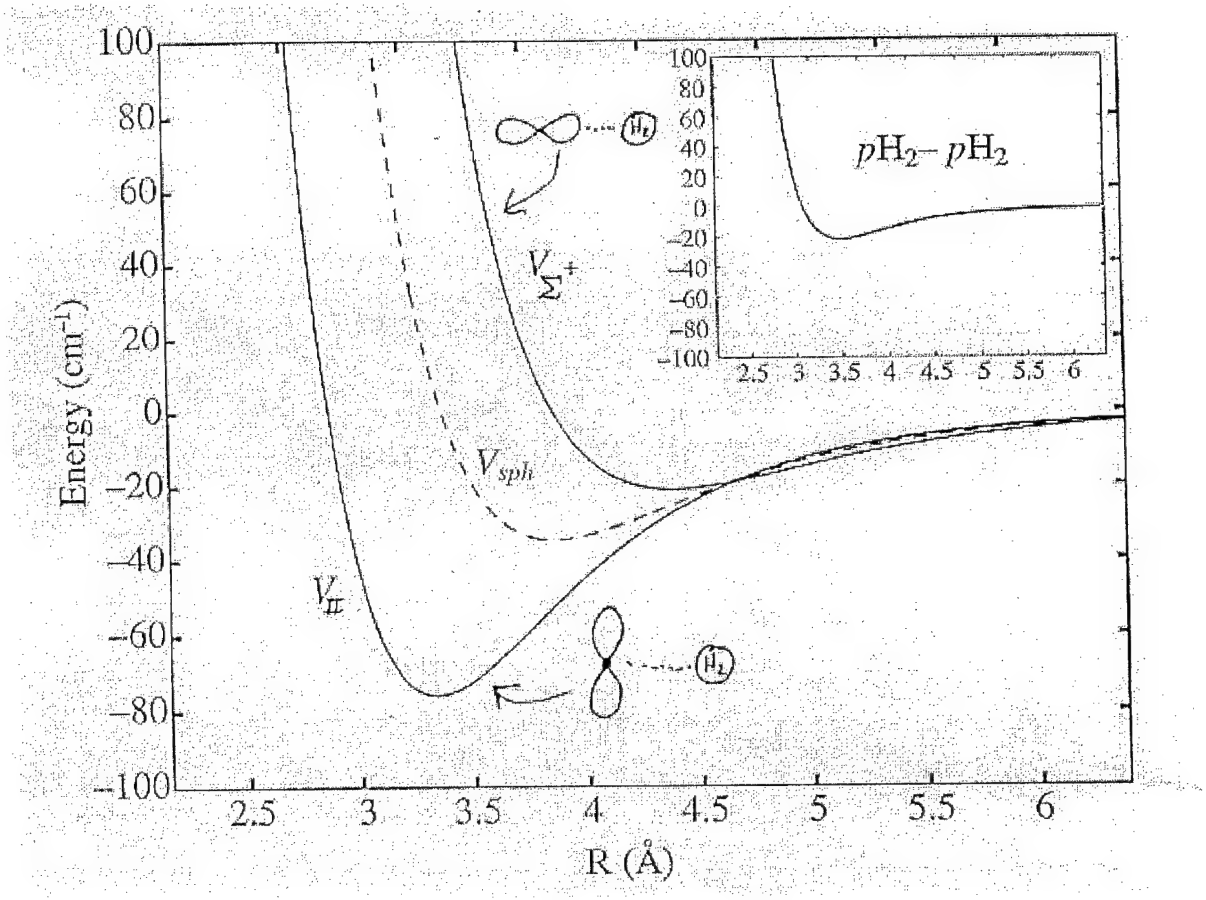
Previous key results

1. In sH_2, B atoms more stable than Li atoms
2. No recombination of B atoms seen at concentrations up to 6.25%.
3. "Forced" recombination of B atoms does not trigger phase separation.

Computational requirements: ~50,000 CPU-hrs, MHPCC IBM SP

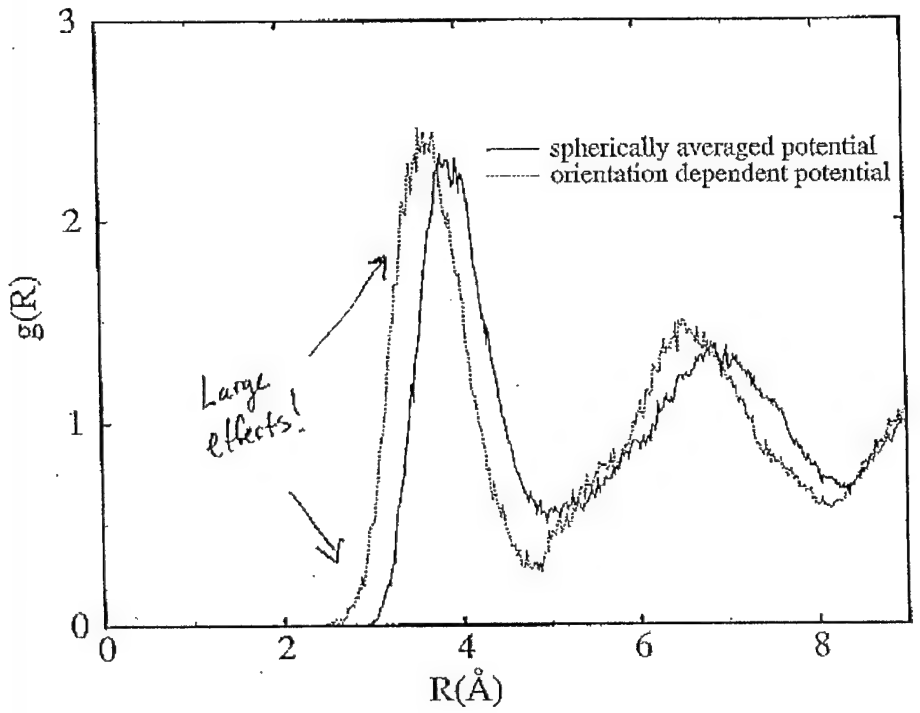
Atom-Doped Solid Hydrogen

How important is the orientational dependence of B-H_2 (Al-H_2) interactions in B/sH_2 (Al/sH_2)?

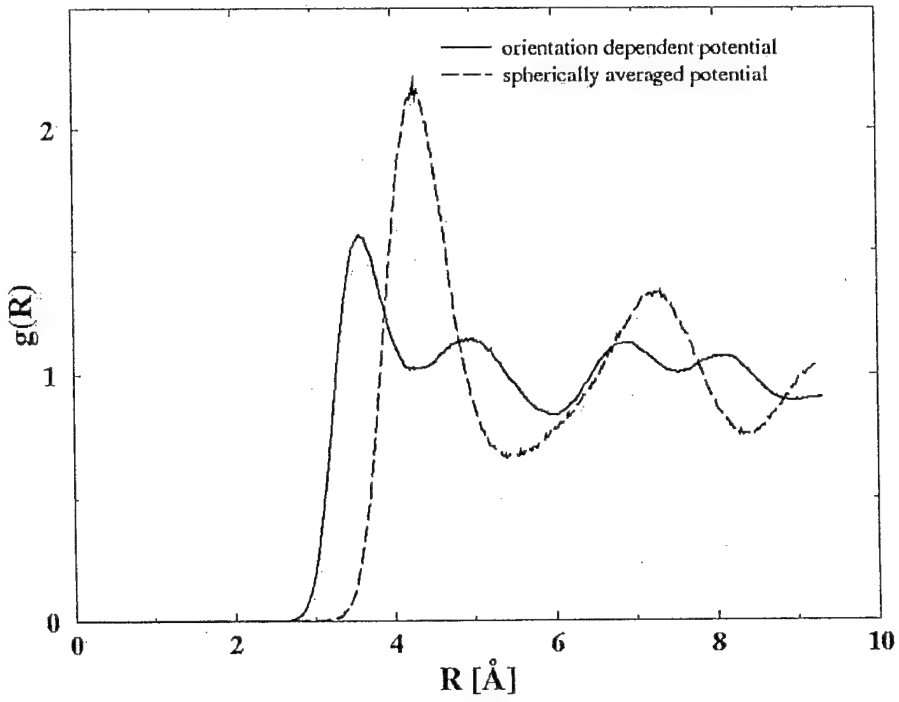


Atom-Doped Solid Hydrogen

B-H_2 pair correlation function



Al-H_2 pair correlation function



Atom-Doped Solid Hydrogen

B-H_2 and Al-H_2 interaction energies (cm^{-1})

		$\langle V_{B\text{-}H_2} \rangle$	$\langle V_{Al\text{-}H_2} \rangle$
Impurity no defect	Orientation dependent	-546.72 (1.92)	-263.41 (3.33)
	Spherically averaged	-443.32 (1.92)	-177.37 (1.26)
Impurity & defect	Orientation dependent	-538.49 (6.06)	-273.86 (2.37)
	Spherically averaged	-439.88 (3.56)	-179.52 (1.90)

Krumrine, J.R., Jang, S., Alexander, M.H., and Voth, G.A.: J. Chem. Phys. 113 (2000) 9079

Mirijanian, D.T., Alexander, M.H., and Voth, G.A.: To be published

RESULTS - POSS

Mechanism of formation

Key steps

1. Hydrolysis of $RSiX_3$ (R=H,CH_3,t-butyl,etc.; X=Cl)

   $RSiCl_3 + H_2O \rightarrow RSiCl_2OH + HCl$

   $RSiCl_2OH + H_2O \rightarrow RSiCl(OH)_2 + HCl$

   $RSiCl(OH)_2 + H_2O \rightarrow RSi(OH)_3$

2. Condensation of $RSi(OH)_3$ to disiloxane

   $2\ RSi(OH)_3 \rightarrow R(OH)_2SiOSi(OH)_2R + H_2O$

3. Condensation of disiloxane to D_3,D_4

   $RSi(OH)_3 + R(OH)_2SiOSi(OH)_2R \rightarrow D_3 + 2H_2O$

   [2+2]: $2R(OH)_2SiOSi(OH)_2R \rightarrow D_4 + 2H_2O$

   [3+1]: $RSi(OH)_3 + R(OH)_2SiOSi(OH)_2R \rightarrow D_4 + 2H_2O$

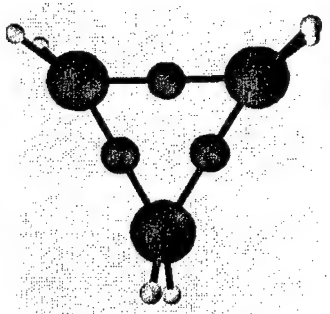
   Ring Expansion: $RSi(OH)_3 + D_3 \rightarrow D_4 + H_2O$

4. Condensation of D_3, D_4 to POSS (in progress)

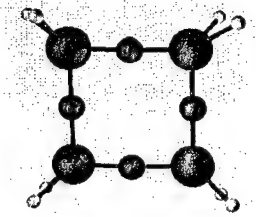
   $2D_4 \rightarrow T_8 + 4H_2O$

   ....

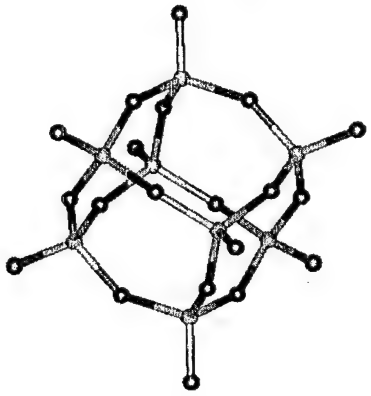
D3



D4



T8



RESULTS - POSS

Mechanism of formation: role of solvent (H_2O) & substituents (R)

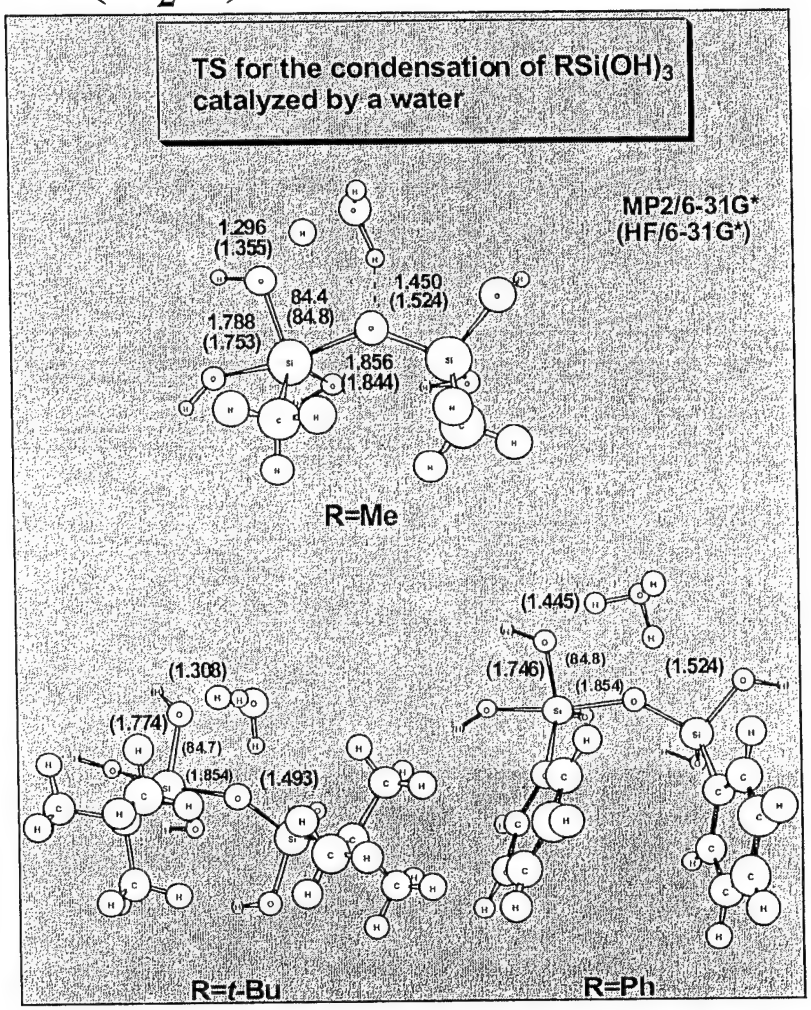
$RSi(OH)_3 + RSi(OH)_3 \rightarrow R(OH)_2SiOSi(OH)_2R + H_2O$

R	Energy barrier (kcal/mol)	
	HF/6-31G*	MP2/6-31G*
H	30.4 (16.7)	10.9 (-9.3)
Me	28.2 (14.7)	7.7 (-13.3)
t-Bu	34.3 (24.9)	9.8 (-9.3)
Ph	31.1 (18.2)	7.9 (-16.4)

Values in parentheses are for water-catalyzed results.

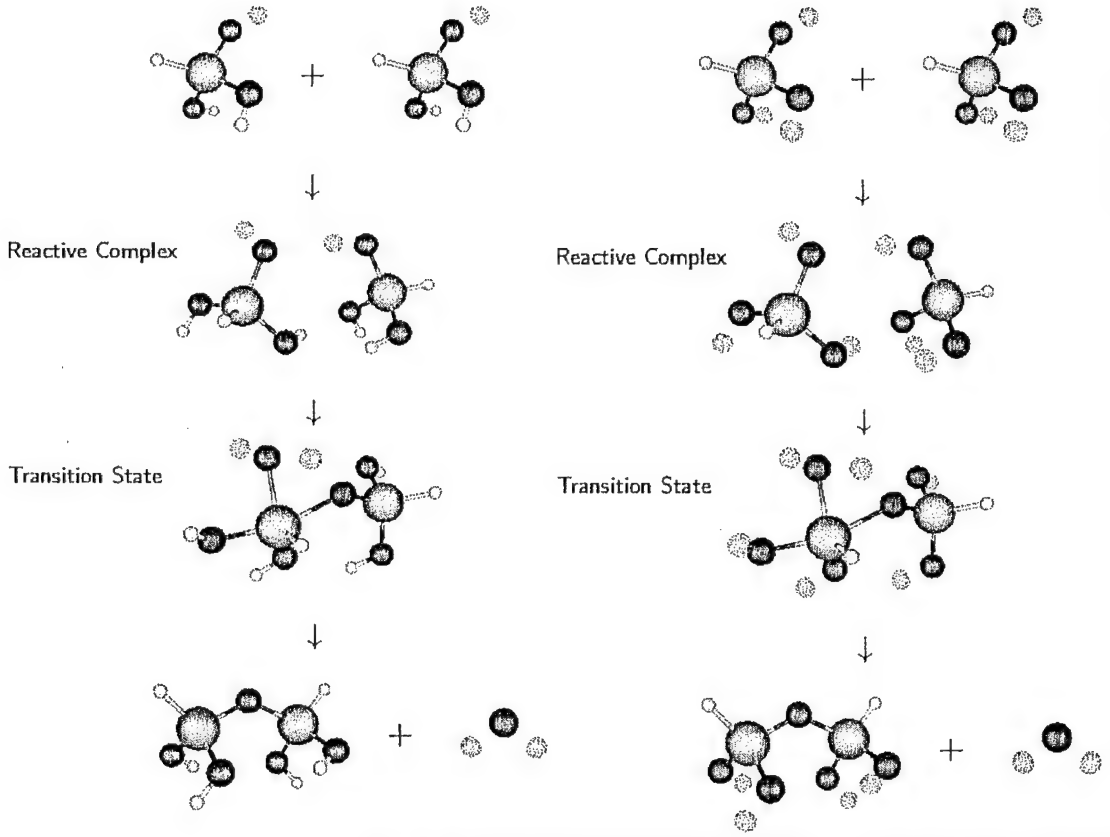
Kudo, T., Gordon, M.S. J. Am. Chem. Soc., 120, 11432 (1998)
Kudo, T., Gordon, M.S. J. Phys. Chem. A, 104, 4058 (2000)

TS for the condensation of $RSi(OH)_3$ catalyzed by a water



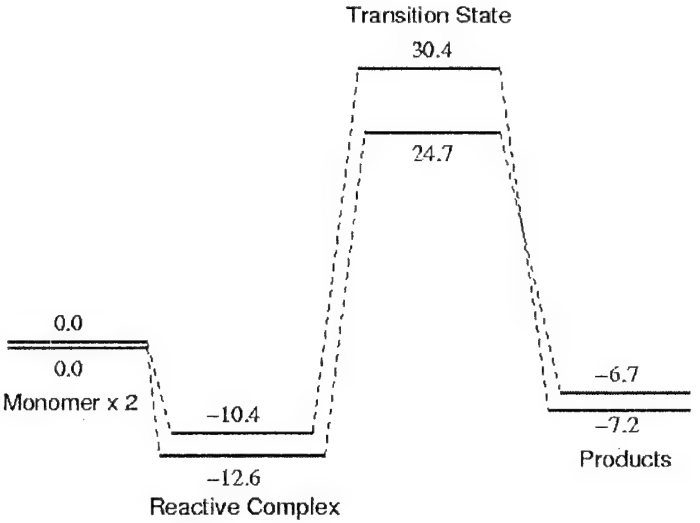
RESULTS - POSS

Nuclear quantum effects in condensation reactions



Level	Monomer × 2	Reactive Complex	Transition State	Products	CPU Time
RHF/6-31G*	0.0	−10.4	30.4	−7.2	1.0
NEO-HF/2	0.0	−11.4	24.9	−5.9	1.1
NEO-HF/6	0.0	−12.6	24.7	−6.7	1.8

Units in kcal/mol



Hammes-Schiffer, S.: J. Phys. Chem. A 102 (1998), 10443

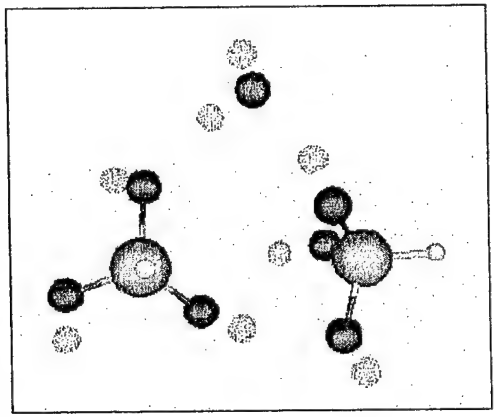
Webb, S.P., Agarwal, P.K., and Hammes-Schiffer, S.: J. Phys. Chem. B, 104(2000), 888

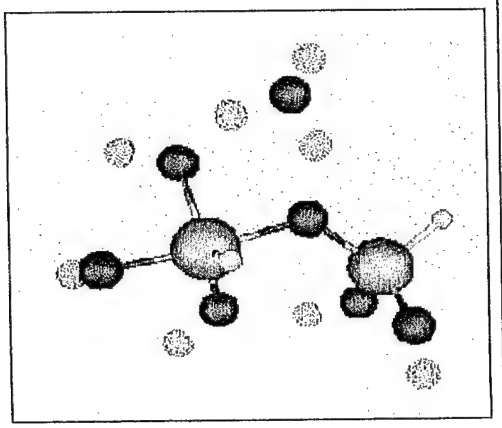
Webb,S.P. and Hammes-Schiffer, S.: J. Chem. Phys. 113 (2000), 5214

RESULTS - POSS

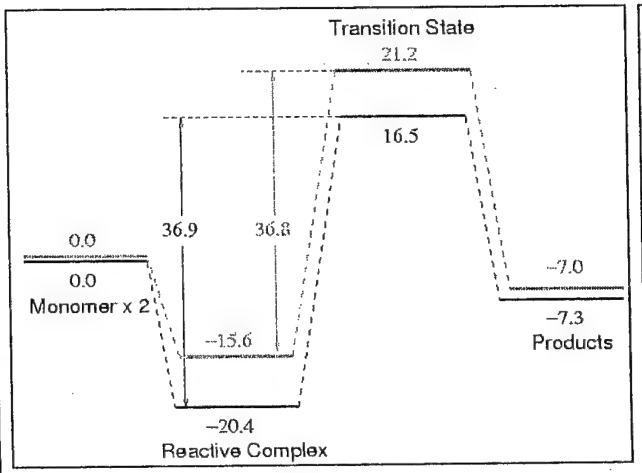
Nuclear quantum effects in water-catalyzed condensation reactions

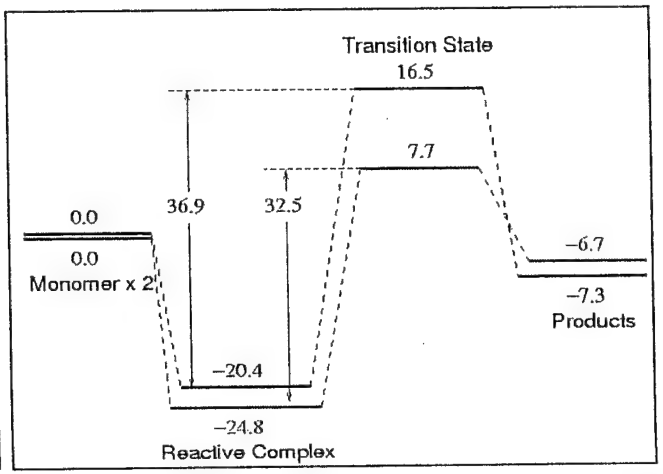
$$2 \times HSi(OH)_3 + H_2O \rightarrow H(OH)_2Si{-}O{-}Si(OH)_2H + 2H_2O$$





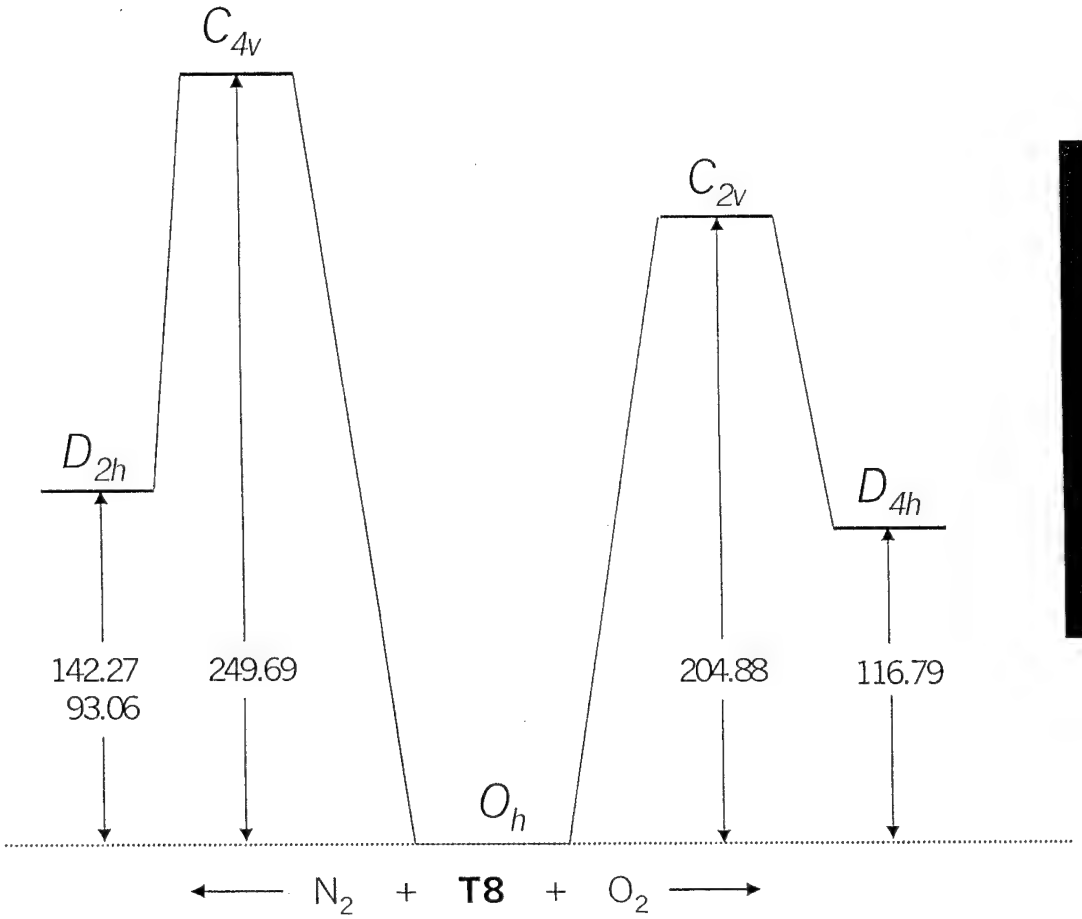
Level	Reactive Complex	Transition State	Products	CPU Time
RHF/6-31G*	–20.4	16.5	–7.3	1.0
RHF/6-31G*/ZPE	–15.6	21.2	–7.0	
NEO/HF/4	–23.0	7.2	–5.9	1.1
NEO/HF/8	–24.8	7.7	–6.7	1.8



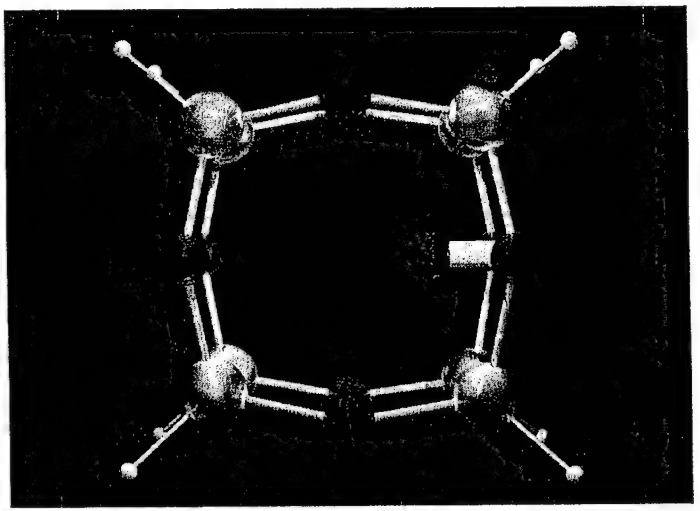


RESULTS - POSS

Molecular "sieves": preferential capture N_2 vs. O_2?



TS structure of $T_8 + O_2$



T_{10} and T_{12} calculations in progress

HPC requirements: ~50,000 node-hrs, AHPCRC T3E, 256 GB

RESULTS - NLO

B3LYP S_0-T_1 Excitation Energies (in eV)

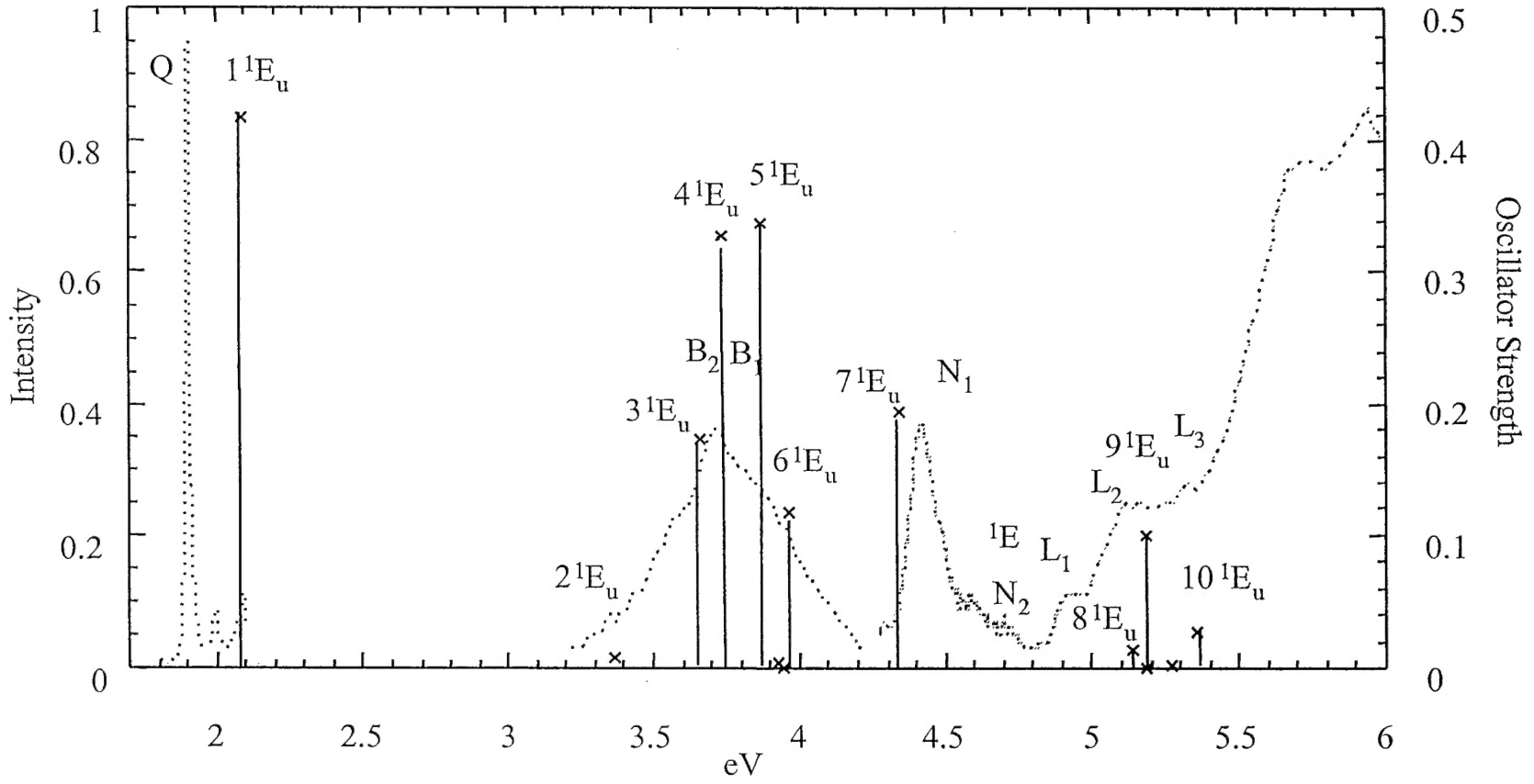
System	6-31G(d)	Error	Exp
Porphyrin (1^3B_{2u})	1.42	0.16	1.58[a]
Zinc Porphyrin (1^3B_{1u})	1.65	0.07	1.72[b]
Tetraphenylporphyrin (1^3B_1)	1.31	0.14	1.45[c]
Zinc Tetraphenylporphyrin(1^3B_1)	1.53	0.06	1.59[d]
Zinc Phthalocyanine (1^3B_{2u})	1.05	0.08	1.13[e]
Zinc Tetrabenzporphyrin (1^3B_{1u})	1.41	0.16	1.57[f]
Phthalocyanine (1^3B_{1u})	1.18	0.06	1.24[g]
Mean Error		0.10	

[a]Gouterman, Khalil, *J. Mol. Spectrosc.* 1974, *53*, 88. (EPA (5:5:2) mixture of ethyl ether to isopentane to ethanol and 50% ethyl iodide at 77 K) [b]Gradyushko, Tsvirko, *Opt. Spectrosc.* 1971, *31*, 291.(EPA at 77 K) [c]Gouterman, Khalil, *J. Mol. Spectrosc.* 1974, *53*, 88. (EPA at 77 K) [d]Walters et al., *J. Phys. Chem.* 1995, *99*, 1166.(1:1 mixture of ether to ethanol at 77 K) [e]Vincett et al.,K. E. *J. Chem. Phys.* 1971, *55*, 4131. (1-chloronapthalene at 77 K) [f]Bajema, Gouterman, *J. Mol. Spectrosc.* 1971, *39*, 421 (octane at 77 K) [g]McVie et al., *J. Chem. Soc. Faraday Trans. II* 1978, *74*, 1870 (1-chloronapthalene at 77 K)

Nguyen, K. A., Day, P. N., and Pachter, R.: J. Chem. Phys,. 110 (1999) 9135
Nguyen, K. A., Day, P. N., and Pachter, R.: J. Phys. Chem. A, 103 (1999) 7378
Nguyen, K. A., Day, P. N., and Pachter, R.: J. Phys. Chem., 104 (2000) 4755
Nguyen, K. A. and Pachter, R., J. Phys. Chem., 104 (2000) 4549

RESULTS - NLO

Comparison with Experiment: ZnPc



HPC Requirements: ~100,000 CPU hours, on SGI O2K + IBM SP3 + SGI O2K @ ASC

SUMMARY

High Energy Density Matter

- High-nitrogen/polynitrogen compounds are more energetic than hydrazine.

- Trityldiazonium cation is not a stable polynitrogen precursor.

- Inclusion of anisotropic interactions of B, Al atoms in sH_2 predict greater stability than spherical interaction model. -

Polyhedral Oligomeric Silsesquioxanes (POSS)

- Proton transfer reactions for hydrolysis and condensation are catalyzed by water.
- Alkyl substituents (R) in $RSiX_3$ have minor effects on hydrolysis and condensation reaction barriers.

- Nuclear quantum effects are important in proton transfer reactions -- lower barriers by >5 kcal/mol.

- T_8 is too small to encapsulate N_2 or O_2.

SUMMARY (cont.)

NLO materials

- Time-dependent density functional theory accurately predicts NLA in porphyrins.
- Computed triplet-triplet excitation energies within 0.1-0.4 eV of experiment
- Computed singlet-triplet excitation energies within 0.1-0.2 eV of experiment
- Computed ionization potentials accurate within 0.1 eV of experiment

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NLO: Kiet Nguyen, Paul Day

GAMESS: Graham Fletcher

MSRCs, DCs: ASC, ARL, ERDC, NAVO, MHPCC, AHPCRC, ARSC, AFFTC

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